Scaling of magnetic fluctuations near a quantum phase transition

A. Schröder[1,2], G. Aeppli[2,3], E. Bucher[4,5], R. Ramazashvili[6], P. Coleman[6]
[1] *Physikalisches Institut, Universität Karlsruhe,D-76128 Karlsruhe, Germany*
[2] *Risø National Laboratory, DK-4000 Roskilde, Denmark*
[3] *NEC, 4 Independence Way, Princeton, NJ 08540, U.S.A.*
[4] *Universität Konstanz, Konstanz, Germany*
[5] *Bell Laboratories, Lucent Technologies, Murray Hill, NJ 079 74, U.S.A.*
[6] *Serin Laboratory, Rutgers University, Piscataway, NJ 08855- 0849*

We use inelastic neutron scattering to measure the magnetic fluctuations in a single crystal of the heavy fermion alloy $CeCu_{5.9}Au_{0.1}$ close to the antiferromagnetic quantum critical point. The energy(E)-, wavevector(q)- and temperature(T)-dependent spectra obey E/T scaling at Q near (1,0,0). The neutron data and earlier bulk susceptibility are consistent with the form $\chi^{-1} \sim f(Q) + (-iE + aT)^{\alpha}$, with an anomalous exponent $\alpha \approx 0.8 \neq 1$. We confirm the earlier observation of quasi-low dimensionality and show how both the magnetic fluctuations and the thermodynamics can be understood in terms of a quantum Lifshitz point.

PACS numbers: 71.27.+a, 75.20.Hr, 75.40.Gb

Quantum phase transitions are zero temperature transitions driven by a parameter such as pressure, magnetic field, or composition which regulates the amplitude of quantum fluctuations. An important realization of the last years has been that many of the most interesting phenomena in condensed matter physics seem to occur near such transitions. Notable examples include high-temperature superconductivity[1,2] and heavy fermion behavior.[3,4] For high-temperature (cuprate) superconductivity, the quantum critical hypothesis leads very naturally to one of the key attributes of the normal state, namely that the energy scale governing spin and charge fluctuations is the temperature T itself, a property labelled E/T scaling.[1]

Heavy fermion behavior is generally understood as a consequence of a competition between intersite spin couplings and the single-site Kondo effect.[5] Antiferromagnetic order develops in a heavy fermion metal at a quantum critical point where the intersite spin couplings overcome the disordering Kondo effect of local spin-exchange processes. While the notion of such competing interactions is old, the nature of the quantum phase transition is a subject of great current interest. $CeCu_6$, a heavy fermion metal with one of the largest known linear specific heats,[3] provides an ideal opportunity to explore this physics in quantitative detail. Here, the quantum critical point (QCP) occurs as Au is substituted for the Cu atoms; when more than 0.1 Cu sites per Ce are replaced, the heavy fermion paramagnet gives way to an ordered antiferromagnet.[3] Fig. 1 illustrates the associated phase diagram and ordering vectors. In spite of the large and rapidly growing literature on this particular QCP[3,6], the E/T scaling which has been so thoroughly tested for the single crystal cuprates[1] and for polycrystalline $UCu_{5-x}Pd_x$[7] has not yet been observed for single crystal heavy fermion systems in general and $CeCu_{6-x}Au_x$ in particular. Our purpose here is to report the discovery, obtained from magnetic neutron scattering, of E/T scaling near the QCP in $CeCu_{5.9}Au_{0.1}$. Beyond establishing the universality of E/T scaling among QCPs hosted by vastly different alloys, our data yield a non-mean-field value for a key critical exponent characterizing the quantum phase transition in $CeCu_{5.9}Au_{0.1}$.

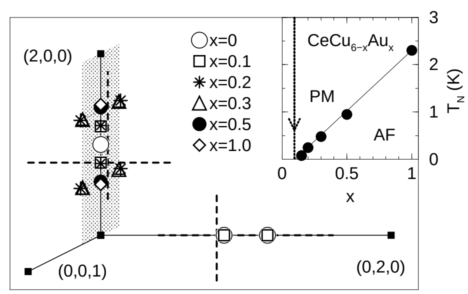

FIG. 1. Location of the magnetic Bragg peaks or spin fluctuation maxima for $CeCu_{6-x}Au_x$ at different Au-concentration x (from refs[9–12]). Dotted lines are the trajectories in the (hk0)-plane of the constant-E scans presented in Fig. 2. Inset shows magnetic ordering temperature T_N (from ref[3]) vs. x. The arrow marks the thermal trajectory investigated for x=0.1 in this paper.

We used the Czochralski technique to grow a single crystal of $CeCu_{5.9}Au_{0.1}$ with volume $\sim$4 cm^3 and a mosaic of 2.5°. To establish the homogeneity and composition of the sample, we used high-resolution neutron diffraction in the orthorhombic [001] zone employed for our inelastic neutron scattering measurements. The result was that throughout the whole sample, the room temperature (Pnma) lattice constants were a = 8.118(2) Å and b = 5.100(1) Å corresponding to the Au concentration of x= 0.10±0.03.[8] Furthermore, the transition into the low-temperature monoclinic phase occurred at a

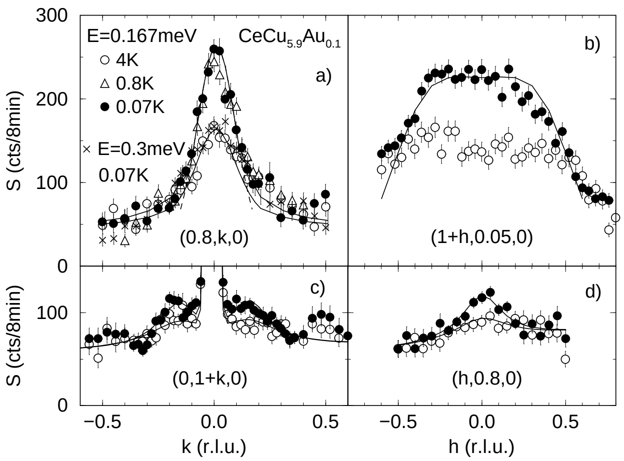

FIG. 2. Constant-E scans through the magnetic scattering from $CeCu_{5.9}Au_{0.1}$ at E_0=0.167 meV near Q=(0.8,0,0) (a,b) and Q=(0 1±0.15 0) (c,d). Solid lines in (a,b) correspond to Eq.(3), corrected for experimental resolution and with the same parameters as shown in Fig. 3 and f(Q) expanded in even powers in Q. The lines in (c,d) are simply guides to the eye. The T-independent peak at (010) in (c) is of nuclear origin.

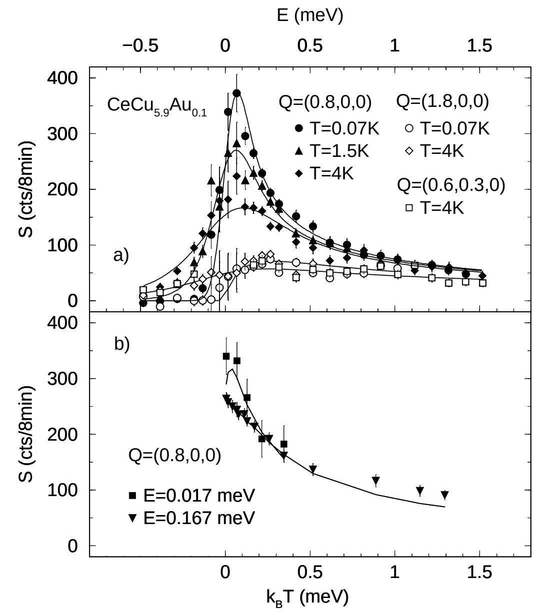

FIG. 3. Constant-Q scans at Q=(0.8 0 0) and Q=(1.8 0 0) and (0.6 0.3 0) at different temperatures T vs energy tranfer E (a) and at different E vs T (b). Lines correspond to Eq(3) with c=83cts/8min·$meV^{0.74}$, where a and α are given by the analysis of the scaled data (see Fig. 4 and text) and include resolution corrections. The optimal (T- and E-independent) values for f(Q) are 0.036(4)$meV^{0.74}$ and 0.38(2)$meV^{0.74}$ for Q=(0.8 0 0) and the other Q-values respectively. Note that f(0.8 0 0) is finite, indicating that our sample is slightly subcritical. But it is sufficiently small that when the data are examined outside the regime where the resolution matters, the deviations from criticality are negligible.

location-independent $T_S = 70\pm2$ K, again confirming the homogeneity of the sample. Between T_S and 5K, the angle γ grows from 90^o to 90.7^o, while the a-axis direction remains unchanged. Thus, the monoclinic distortion is sufficiently small that we follow past custom and use orthorhombic notation to label points in reciprocal space.

After installing the sample in the Bell Laboratories/Risø dilution refrigerator, we collected inelastic neutron scattering data using TAS7 at the Risø DR3 reactor equipped with a PG(002) monochromator and analyzer and a BeO filter. The final neutron energy was fixed at E_f= 3.7 meV. The energy resolution, defined as the full-width-at-half-maximum (FWHM) of the elastic incoherent scattering from the sample, was 0.134 meV.

When we began our experiments, we knew that the most intense magnetic fluctuations in the paramagnetic parent $CeCu_6$ were located close to the inequivalent reciprocal lattice points (010) and (100).[9] At the same time, the incommensurate vectors (1±δ 0 0) fully describe the magnetic order in $CeCu_{6-x}Au_x$ with x≥0.5[10,11] and do so partially for x= 0.2.[12] Thus, to discover the dominant fluctuations near the QCP at x= 0.1, we investigated fluctuations with wave-vectors of both types. Fig. 2 shows the corresponding constant-energy scans at E_0= 0.167meV, collected along the trajectories indicated in Fig. 1. Frames (c) and (d) demonstrate that incommensurate peaks survive at (0 1±0.15 0), and that their intensity grows upon cooling. The widths of the (0 1±0.15 0) peaks in the (1,0,0) and (0,1,0) directions are similar and not resolution-limited, corresponding to magnetic correlations with a range of order 8Å(at this E_0). Even though they are still present and growing with decreasing T, the (0 1±0.15 0) peaks are five times weaker than the scattering, shown in frames (a) and (b), near (100). Here, there is a weak modulation along (1,0,0) with hints of maxima at (1±δ 0 0) for $\delta \approx 0.2$, and a much sharper modulation along (0,1,0). Both the peak intensity and width along (0,1,0) are clearly T and E-dependent, corresponding to growing antiferromagnetic correlations with decreasing T and E. Furthermore, T and E appear interchangeable in the sense that the profiles for E=0.3meV > k_BT=0.006meV and E=0.167meV < k_BT=0.345meV are indistinguishable. That the larger of E and k_BT controls the width κ(E,T) is expected for quantum critical points and has been previously observed in the context of high-temperature superconductors.[2]

We next characterize the spectrum of the most intense fluctuations, namely those at Q=(1±0.2 0 0). Fig. 3 shows such constant-Q spectra for several T. For comparison, it also shows data at Q=(1.8 0 0) and (0.6 0.3 0), for which no special Q- or T-dependent enhancement is found. The Q- and T-independent elastic scattering background has been subtracted from all data shown. At Q=(0.8 0 0), the spectra narrow dramatically with reduction in T, to the point where they have a rising edge at low E which is comparable to that of an instrumentally broadened step function. Thus, as happens near any second order phase transition, cooling results in a reduced

magnetic relaxation rate. Even so, a long high-E tail of the spectra persists at all T that the FWHM never approaches that of the instrumental resolution. At Q=(1.8 0 0), the spectra change much less dramatically.

In addition to performing measurements as a function of E at fixed T, we have also collected data as a function of T for fixed E (Fig. 3). The similarity between the T-scans (b) and the E-scans (a) suggests E/T scaling, which we have tested by fitting the data to the form

$$\chi''(E,T) = T^{-\alpha} g(E/k_B T). \tag{1}$$

No assumptions were made about $g(x)$ beyond the ability to approximate it by a histogram with stepsize 0.1 on a $\log_{10}$ E/T scale between -0.5 and 0.7. The best collapse of the data onto a single curve, as checked by the smallest (log) deviation ($\sigma_{log(g)}$ see inset) from the mean step values, is obtained for $\alpha = 0.75 \pm 0.1$ and shown in Fig. 4, which displays all the data at Q=(0.8 0 0) unaffected by the finite experimental resolution.

Having confirmed E/T scaling, we ask whether the associated exponent α is consistent with other data and theory. The simplest mean field approach[13] dictates a susceptibility χ

$$\chi(Q,E,T)^{-1} = c^{-1}(f(Q) - iE + aT), \tag{2}$$

where f(Q) is a smooth function of Q with minima at the wave-vectors characterizing the eventual magnetic order. If we position ourselves at the critical Q and composition, f(Q)=0 and $\chi'' = cE/[(aT)^2 + E^2]$, which we can compare to Eq.(1) to read off the scaling function $g_o(y) = y/(1+y^2)$ with $y = E/aT$ and the exponent α=1. The dotted line in Fig. 4(a) corresponds to the (log) optimized $g_o(y)$ with a/k_B=1 and deviates clearly from the data.

Neither $g_o(y)$ nor α= 1 account for the measurements; thus the simple mean-field theory fails for $CeCu_{5.9}Au_{0.1}$. To make progress, we modify Eq(2) to account for arbitrary values of α via the simple expedient of replacing $(-iE + aT)$ in Eq.(2) by $(-iE + aT)^\alpha$,

$$\chi^{-1} = c^{-1}(f(Q) + (-iE + aT)^\alpha) \tag{3}$$

Beyond accounting for the anomalous value of the exponent of T in the scaling relation, this generalization has two other important implications. The first is that the scaling function (for Q at which f(Q)= 0) becomes

$$g(y) = c \sin[\alpha \tan^{-1}(y)]/(y^2+1)^{\alpha/2} \tag{4}$$

The solid line in Fig. 4 corresponds to the best fit of Eq.(4) to the data (with c=88cts/8min$\cdot$meV$^{0.74}$); note that the value obtained from floating α in Eq.(4) in the fitting process is at 0.74±0.1 indistinguishable from that obtained as the exponent for the thermal prefactor α in Eq.(1). At the same time, a/k_B=0.82 is a number close to unity, indicating that the magnetic fluctuations have an underlying energy scale very close to the thermal energy.

The second consequence of the generalized form (3) is that for E=0, $\chi'(Q,T)^{-1} = c^{-1}(f(Q) + (aT)^\alpha)$, which

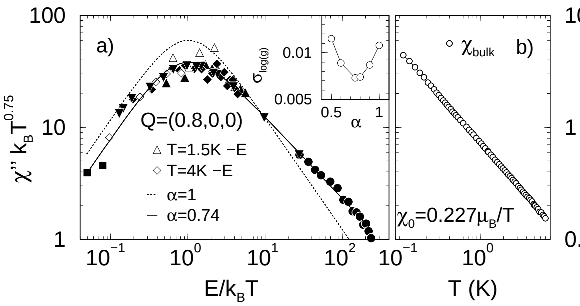

FIG. 4. (a) Scaling plot of the data from Fig. 3 for Q=(0.8 0 0). The inset shows how the quality of the scaling collapse varies with α in Eq.(3), using a procedure unbiased towards any particular form of the scaling function g(y) (see text). Solid and dashed lines correspond to Eq.(4) with α=0.74 and 1, respectively. (b) Double logarithmic plot of susceptibility χ_{bulk} (from M/B in B_c =0.1T from[3]) vs. T after subtracting a zero T value χ_0 corresponding to a 'parallel' shunt, as suggested by Eq.(3).

implies that $(\chi'(Q,T)^{-1} - \chi'(Q,0)^{-1})^{-1} = c(aT)^{-\alpha}$.We test this relation by using the very accurate and dense data provided by bulk magnetometry[3] for a special value of Q, namely Q=0. The fact that on the double logarithmic scale of Fig. 4(b) the data lie on a straight line shows that the relation is indeed satisfied. The line is not parallel to the diagonal for which α=1, but indeed has slope α=0.8±0.1, indistinguishable from α obtained from the neutron scattering data by the other two methods described above.

The neutron results are consistent with the bulk susceptibility: do they also account for the thermodynamics? One of the key features of this system, is that, at low T, $C/T = \gamma(T)$ diverges logarithmically.[3] To see whether $\gamma(T) \sim \ln(T)$ is consistent with the neutron measurements, we need to consider the detailed dependence of $f(Q)$. In keeping with earlier investigations[6], the critical fluctuations are almost two-dimensional. Specifically, the constant E scans along (1,0,0) (Fig. 2(b)) look flat-topped, indicating that the quadratic term in the power series expansion of $f(Q)$ around (100) almost vanishes in this direction. A good fit to the data in the vicinity of $Q = (0.800)$ is given by $f((100)+\delta Q) = (0.036 + 12(\delta k)^2 - 0.08(\delta h)^2 + 2(\delta h)^4) meV^{0.74}$ (see dashed/solid lines in Fig. 2). That the quadratic term $B_h(\delta h)^2$ is negative (with a positive quartic term $C_h(\delta h)^4$) also follows from the observation that $CeCu_{6-x}Au_x$ orders not at (100) but at pairs of incommensurate wavevectors nearby (see Fig. 1). As the sign of B_h changes, the minimum at δQ = 0 of f(δQ) splits into the pair $\delta Q = \pm\sqrt{-B_h/2C_h}$. In the classical theory of phase transitions, vanishing of the quadratic stiffness occurs at a Lifshitz point[14]. Here, we are dealing with a quantum Lifshitz point.

To understand the thermodynamics of a quantum Lifshitz point, we assume that the quadratic stiffness vanishes along only one direction (e.g.(1,0,0)) in reciprocal

space or more generally, that it vanishes for a finite number of lines centered at certain points Q in reciprocal space[14]. The most important contributions to χ will then be proportional to $1/[B_\perp(\delta Q_\perp)^2 + C_\parallel(\delta Q_\parallel)^4 + (-iE + aT)^{\frac{2}{z}}]$ in the vicinity of Q, where $\delta Q_\parallel$ and $\delta Q_\perp$ measure the departure from Q parallel and perpendicular to the line direction, and we have employed the notation of dynamical critical phenomena, writing $\alpha = 2/z$. If $\kappa_\perp$ and $\kappa_\parallel$ are the inverse correlation lengths in these directions

$$\kappa_\perp \sim T^{1/z}, \qquad \kappa_\parallel \sim T^{1/z_\parallel} = T^{1/2z},$$

so the dynamical critical exponent is $2z$ along the $(1,0,0)$ direction, but z in the perpendicular directions. This has the effect of reducing the effective dimensionality D_{eff}. For an isotropic QCP, the free energy scales as $F \sim T\kappa^D \sim T^{1+D/z}$. For a quantum Lifshitz point, we must now write

$$F \sim T\kappa_\perp^{D-1}\kappa_\parallel \sim T^{1+(D-\frac{1}{2})/z}.$$

So D_{eff} is lower by *one half*, and

$$\gamma(T) = -\partial_T^2 F(T) \sim T^{(D-\frac{1}{2})\alpha/2-1}.$$

We see that for $D = 3$, $\alpha = 0.8$, the exponent for $\gamma(T)$ vanishes, in accord with the specific heat data. If we ignore the anisotropic form of f(Q) indicated by the neutron scans through reciprocal space and assume a conventional (non-Lifshitz) QCP for $CeCu_{6-x}Au_x$, the α obtained from the energy- and temperature-dependence of the neutron data as well as the bulk χ would lead to $\gamma \sim T^{0.2}$ and $T^{-0.2}$ for D=3 and 2 respectively, in disagreement with the C(T) results.

Our ability to use the phenomenological scaling form for χ^{-1} for both the thermodynamics *and* the uniform susceptibility indicates that the anomalous energy dependence of the susceptibility extends over a wide region of the Brillouin zone. We can write the T=0 susceptibility in (3) as $\chi^{-1}(Q,E) = \chi^{-1}(E) - J(Q)$, where $\chi(E)$ describes the response of individual local moments and surrounding conduction electrons and $J(Q)$ defines the interactions between different screened local moments. Eq.(3) actually determines $\chi^{-1}(E)$ and $J(Q)$ up to the same additive constant Θ which sets the scale for the screening: $\chi^{-1}(E) = c^{-1}(-iE)^\alpha + \Theta$ and $J(Q) = -c^{-1}f(Q) + \Theta$. Our result that $\alpha = 0.8 \pm 0.1$ implies that $\chi(E)$ is nonanalytic near E=0. In the "Millis-Hertz" theory of a quantum critical point, the soft magnetic fluctuations couple to a Fermi liquid and the corresponding local response is analytic, with $\alpha = 1$. The anomalous exponent $\alpha \neq 1$ for $CeCu_{5.9}Au_{0.1}$ thus represents a fundamental *local* deviation from Fermi liquid behavior. One possible interpretation is that the magnetic QCP has qualitatively modified the underlying moment compensation mechanism,[15] causing the local electron propagator to develop an anomalous scaling dimension.[16]

We have performed the first detailed single crystal measurements of the magnetic fluctuation spectrum in a heavy fermion system close to an antiferromagnetic quantum critical point. We find that the data obey E/T scaling with an anomalous scaling exponent $\alpha \approx 0.8$. The same exponent appears in three other independent quantities: the scaling function itself, previously published bulk susceptibility,[3] and the specific heat. To obtain the scaling relation connecting C(T) with the magnetic measurement, we take into account the (again independent) indications of our neutron data that the QCP in $CeCu_{5.9}Au_{0.1}$ is actually near a quantum Lifshitz point, whose hallmark is the near degeneracy of ordered states with characteristic wavevectors along some line(s) in reciprocal space. The general idea that there are many potential ground states for $CeCu_{5.9}Au_{0.1}$ is further reinforced by our observation of an enhanced susceptibility at the seemingly inequivalent locations (0 1±0.15 0) and (1±0.2 0 0) in reciprocal space.

We gratefully acknowledge discussions with G. Kotliar, G. L. Lonzarich, H. v. Löhneysen, A. Rosch and O. Stockert. Research at Karlsruhe was supported by the Deutsche Forschungsgemeinschaft. Research at Rutgers was supported in part by the National Science Foundation under grant NSF DMR 96-14999.

---

[1] B. Keimer *et al.* Phys. Rev. Lett. **67**, 1930 (1991); S M. Hayden *et al.* Ibid. **66**, 821 (1991); C.M. Varma *et al.* Ibid. **63**, 1996 (1989); S. Sachdev and J. Ye Ibid. **69**, 2411 (1992).

[2] G. Aeppli *et al.*, Science **278**, 1432 (1997).

[3] H. v. Löhneysen *et al.*, Phys. Rev. Lett. **72**, 3262 (1994), H. v. Löhneysen, J. Phys. Cond. Matter **8**, 9689 (1996) and refs therein,

[4] e.g. S.J.S. Lister *et al.*, Z. Phys. B **103**, 263 (1997)

[5] S. Doniach, Physica B **91**, 231 (1977); G. Aeppli and C. Broholm p.123 in Vol 19, Handbook on the Physics and Chemistry of Rare Earths (ed. by Gschneidner *et al.*, Elsevier, 1994)

[6] A. Rosch *et al.*, Phys. Rev. Lett. **79**, 159 (1997)

[7] M. C. Aronson *et al.* Phys. Rev. Lett. **75**, 725 (1995)

[8] H. G. Schlager *et al.* , J. Low Temp Phys. **90**, 181 (1993)

[9] G. Aeppli *et al.*, Phys. Rev. Lett. **57**, 122 (1986); J. Rossat-Mignod *et al.*, J. Magn. Magn. Mater. **76-77**, 376 (1988)

[10] H. v. Löhneysen *et al.*, J. Magn. Magn. Mater., to be published; H. Okumura *et al.*, *ibid.* ,to be published; O. Stockert *et al.*, to be published

[11] A. Schröder *et al.*, Physica B **199-200**, 47 (1994)

[12] O. Stockert *et al.*, Physica B **230-232**, 247 (1997)

[13] John A. Hertz, Phys. Rev. **B 14**, 1165 (1976); A. J. Millis, Phys. Rev. B **48**, 7183 (1993).

[14] see, e.g., P.M. Chaikin and T.C. Lubensky, "Principles of condensed matter physics",p. 184, Cambridge University Press, 1995

[15] A. I. Larkin and V. I. Melnikov, JETP **34**, 656 (1972).

[16] P. W. Anderson, Phys. Rev. Lett. **67**, 2092, (1991).